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THE COLLISIONAL DE-EXCITATION OF
NEON AT VARIOUS PRESSURES

Ivan Robert Farris

NAVAL POSTGRADUATE SCHOOL

Monterey, California



THESIS

THE COLLISIONAL DE-EXCITATION
OF
NEON AT VARIOUS PRESSURES

by

Ivan Robert Farris

Thesis Advisor:

E. A. Milne

June 1972

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The Collisional De-excitation
of
Neon at Various Pressures

by

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Submitted in partial fulfillment of the
requirements for the degree of

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June 1972

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ABSTRACT

It was proposed to observe the spectral intensities of the 5852.5Å^o line in Neon I, and 3664.1Å^o line in Neon II to obtain collisional de-excitation cross sections, σ_d , and de-excitation reaction rates, K_i .

However, due to a series of problems with the Van De Graaff accelerator in the proton configuration no reproducible data was obtained. The majority of the time was thus spent converting the accelerator to the electron configuration and attempting to make the system operational again. In this light trial runs were made on the 3371.3Å^o spectral line of nitrogen using electrons as the excitation source. Values obtained in these runs were off by a factor of one thousand from those obtained by A. R. Smelley [Ref. 4] using protons.

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I. INTRODUCTION

Prior to this work studies have been made on argon and nitrogen, here at the Naval Postgraduate School. In this thesis it was proposed to continue this general line of studies. More specifically the $2p^5 3s - 2p^5 3p$ transition in Ne I ($\lambda = 5852.49$) and the $2p^4 3s - 2p^4 3p$ transition in Ne II ($\lambda = 3664.11$) were to be observed. Target chamber pressures were to range from 10 to approximately 700 torr. A second part of the experiment was to observe the quenching effects of N_2 gas on Neon. This would be accomplished by admitting Neon to the target chamber first (up to 100 torr) and then adding N_2 while holding the Neon partial pressure constant. From a plot of pressure divided by relative intensity versus pressure, the collisional de-excitation rates and collisional de-excitation cross sections for the two proposed spectral lines will be determined. Originally it was planned to use 1.5 Mev protons to excite the Neon gas, but due to uncorrectable trouble in the accelerator tube, the Van De Graaff accelerator was converted to an electron accelerator. Due to a shortage of time and "bugs" encountered in the switchover, no reproducible/confirmatory data was obtained.

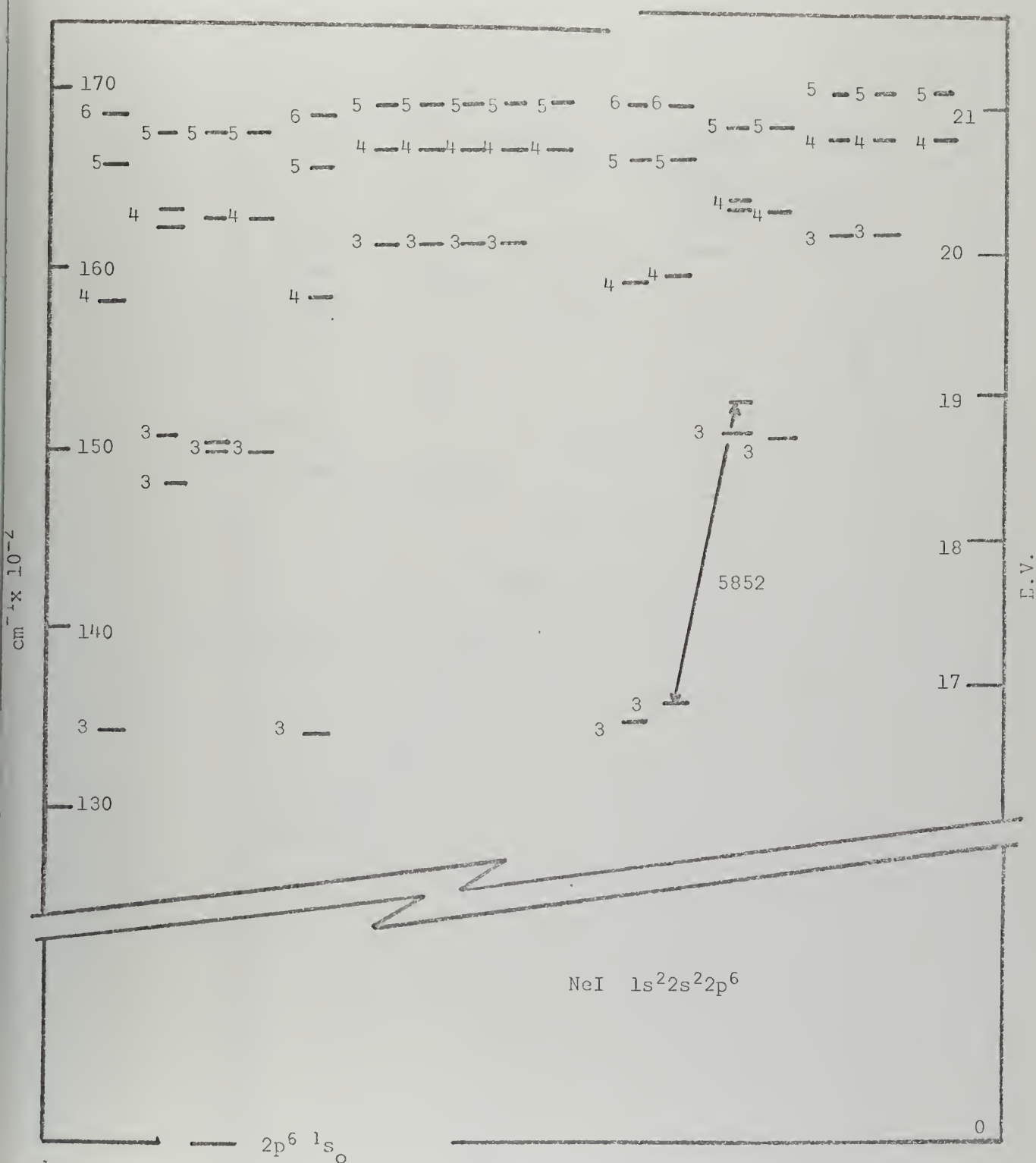


Figure 1. Energy Level Diagram for Neon I

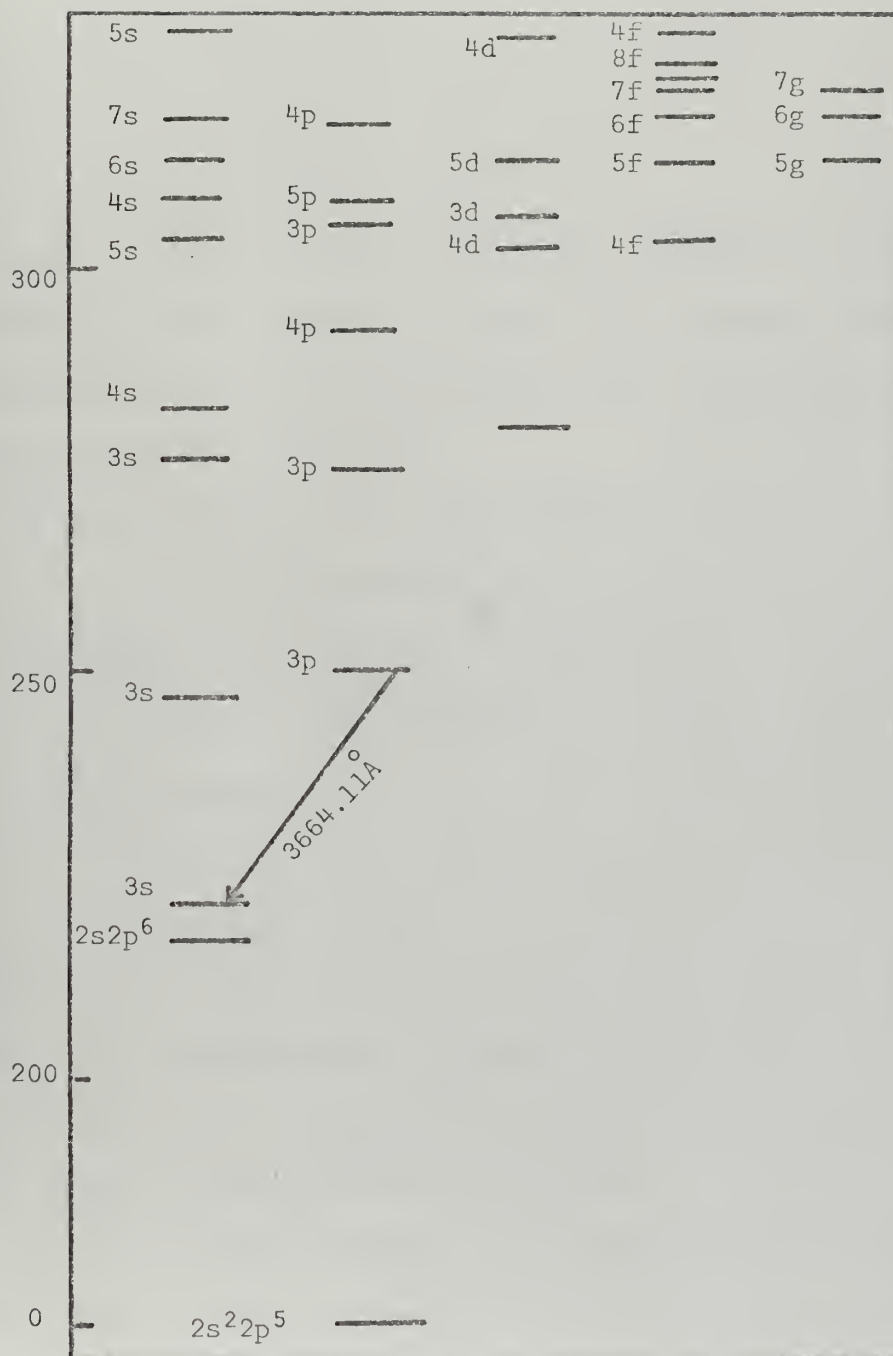
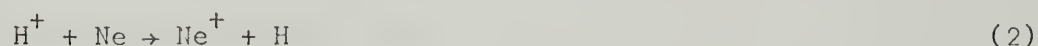
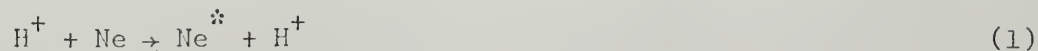


Figure 2. Energy Level Diagram for Neon II

II. THEORY

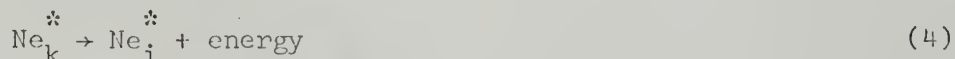
Neon may be excited by the following methods:

a) By protons:

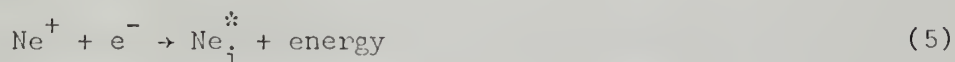


Equation (1) represents direct excitation. Equation (2) represents excitation by charge transfer. Equation (3) represents excitation by simultaneous ionization and excitation. Two other possibilities exist for getting Neon in *i*th state:

By cascade from a higher excited state.



By electron capture from an ionized state.

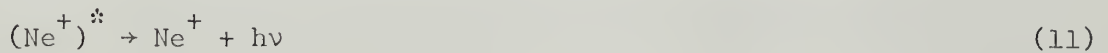
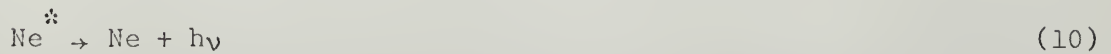
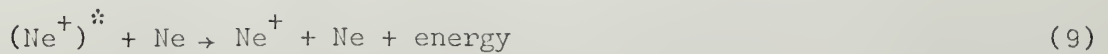
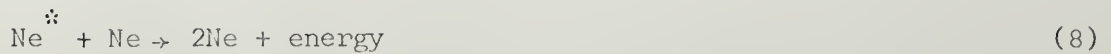


b) By electrons:



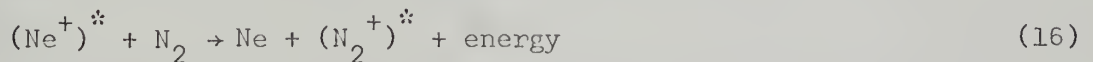
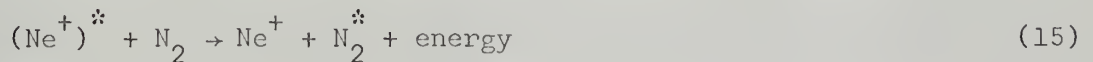
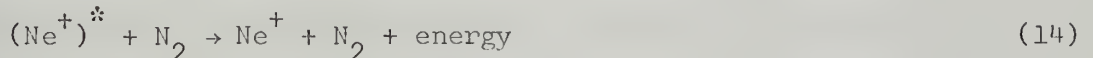
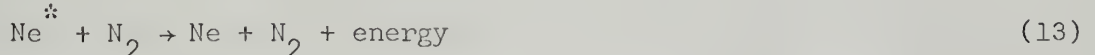
Equation (6) is direct excitation by electrons. Equation (7) is excitation by simultaneous ionization and excitation. Equations (4) and (5) would also be applicable for getting Neon into the *i*th excited state when using electrons for excitation.

Having thus raised the Neon to the *i*th excited state by any of the above methods (protons or electrons) the possibilities for de-excitation are:



where Eqs. (8) and (9) are collisional de-excitations, and Eqs. (10) and (11) are radiative de-excitations.

When N_2 is used as a quenching agent there will be additional de-excitation possibilities, in addition to Eqs. (8) - (11). The additional possibilities are:



Given a beam (protons or electrons) with current density equal to J , and incident upon the neon target gas, whose density is N atoms/cm³, the beam yields a population of an excited state i of the atomic (neon in this case) species L with respect to time expressed by

$$\begin{aligned} \frac{dN}{dt} \text{Li} = & \frac{J\sigma_{\text{Li}}}{e} N_{\text{Lg}} + \sum_{\substack{j \\ j>i}} \lambda_{\text{Lji}} N_{\text{Lj}} - \sum_{\substack{j \\ j<i}} \lambda_{\text{Lij}} N_{\text{Li}} + \sum_{m,k} K_{\text{kiLm}} N_{\text{Lg}} N_{\text{mk}} \\ & - \sum_m K_{\text{igLm}} N_{\text{Li}} N_{\text{mg}} \end{aligned} \quad (17)$$

The subscripts denote the following:

L = particular molecular species being observed (neon)

m = any atomic species

g = ground state

j, k = any other state

i = excited state being observed

λ_{Lji} = transition probability for species L from excited state j to excited state i

K_{igLm} = collisional rate coefficient for de-excitation of excited state i in species L by collisions with molecules of species m in the ground state

The first term in Eq. (17) indicates the rate of population of the excited state being observed, excited state i , due to direct excitation of the ground state by proton, or electron, impact.

The second term represents the rate of population of excited state i through radiated transitions from higher excited states, while the third term represents the de-population of excited state i by radiated transitions to lower excited states or the ground state.

The fourth term is the rate of population of excited state i from collisions of atoms of the observed species with atoms or molecules, whichever the case may be, of species type m in the state k . The fifth and last term indicates the de-population resulting from collisions between atoms in state i , the excited state being observed for the species under analysis, and atoms of any other species m , the atoms of species m being in the ground state.

The following assumptions were made to get Eq. (17) into a manageable form.

- | | |
|----------------------------------|--|
| 1) $P = NKT$ | Perfect gas law holds for the target gas. |
| 2) $\frac{J}{e} \sigma_{Li} = d$ | A constant for each line observed. |
| 3) $N_{Lg} \propto P_L$ | The density of the ground state of the target gas is proportional to the partial pressure of the target gas. |

- 4) $\sum_{j>i} \lambda_{Lji} N_{Lj} = f P_L$ Population of the i th state from radiated transitions of a higher state is proportional to the partial pressure of the target gas.
- 5) $\sum_{j<i} \lambda_{Lij} N_{Li} = \lambda_{Li} b I$ Population of the excited state i is proportional to the observed spectral line intensity.

The assumption of the perfect gas law is valid for the pressure ranges used in this study [Ref. 10]. Also, since the excitation cross section σ_{Li} is energy dependent, and target gas density, current density and energy are to be constant for each data point, this cross section is constant for each data point. $\sum_{j<i} \lambda_{Lij}$ in assumption 5) above is constant for any given spectral line and therefore represented by λ_{Li} .

Utilizing the above assumptions and assuming equilibrium conditions transforms Eq. (16) into

$$\frac{dN_{Li}}{dt} = 0 = a P_L - \lambda_{Li} b I + \sum_{m,k} K_{kiLm} \frac{P_L N_{mk}}{\kappa T} - \sum K_{igLm} \frac{b I P_m}{\kappa T} \quad (18)$$

κ is the Boltzmann constant, T is the temperature and a and b are proportionality constants.

For neon as the target gas with no quenching nitrogen gas, Eq. (18) becomes:

$$0 = aP - \lambda_i b I - \frac{K_i b I P}{\kappa T} \quad (19)$$

Which can be written as:

$$I = \frac{P}{A + BP} \quad (20)$$

where $A = \frac{\lambda_i b}{a}$ and $B = \frac{K_i b}{a \kappa T}$.

By inspection of Eq. (20) an experimental plot of P/I versus P should yield a straight line of slope equal to B and intercept equal to A. The ratio of B over A is:

$$B/A = \frac{K_i}{\lambda_i \kappa T} \quad (21)$$

Therefore if the transition probability λ_i is known for the observed spectral line, K_i , the collisional de-excitation rate coefficient can be computed. λ_i for spectral line 5852.49 $\overset{\circ}{\text{\AA}}$ is taken to be $0.719 \times 10^8 \text{ sec}^{-1}$ from Ref. 8. λ_i for spectral line 3664.11 $\overset{\circ}{\text{\AA}}$ is taken to be $0.67 \times 10^8 \text{ sec}^{-1}$ again from Ref. 8. T, the temperature was assumed to be approximately 300 $^\circ$ Kelvin.

For neon as the target gas with nitrogen as a quenching agent Eq. (18) becomes:

$$0 = aP_L - \lambda_{Li} bI - K_{igLm} \frac{bIP_m}{\kappa T} - K_{igLL} \frac{bIP_L}{\kappa T} \quad (22)$$

where the energies of the excited states of the quenching gas are less than the energy of the atoms of the target gas in excited state i. This is due to the fact that neon is a noble gas and as such its excited state energy levels are above those of the non-noble quenching gas, nitrogen. Thus there is de-excitation of neon in state i due to collisions with molecules of the quenching gas but no excitation of the neon by the quenching gas. There is also a de-excitation of the neon in state i due to collisions with lower energy level neon atoms. Equation (22) can be manipulated as was previously done with Eq. (19) to give:

$$I = \frac{P_L}{A + BP_L + CP_m} \quad (23)$$

With: P_m = Pressure due to nitrogen (quenching agent)

P_L = Pressure due to neon (target gas)

$$A = \frac{\lambda_{Li} b}{a}$$

$$B = \frac{K_{igLL} b}{a\kappa T}$$

$$C = \frac{K_{igLm} b}{a\kappa T}$$

K_{iglm} is the rate coefficient for the N_2 quenching gas. The constants A and B above are determined as before with the stopping point being the desired partial pressure. After determination of A and B is made the pressure of the target gas (neon) is held constant and the quenching gas is admitted to the chamber in increments. A plot of P_L/I versus P_m then yields a straight line with slope equal to C and intercept equal to $A + BP_L$. Then from

$$\frac{C}{A + BP_L} = \frac{K_{igLm}}{\lambda_{Li}\kappa T + K_{igLL} P_L} \quad (24)$$

a determination of K_{igLm} , the rate coefficient of the quenching gas, can be made.

III. EXPERIMENTAL PROCEDURE

Figure 3 depicts the experimental set up with associated electronic equipment. A beam of 2 Mev protons or 1 Mev electrons from a Van De Graaff generator is used to excite the target gas. Vacuum in the accelerator tube is approximately 10^{-6} torr with the target chamber separated from the tube by an aluminum foil window. Foil windows are 1/2 mill when using protons and 1 mill when using electrons.

The target chamber itself is a glass (Pyrex) tee which can be evacuated to a pressure of approximately 4×10^{-7} torr using a liquid nitrogen trapped oil diffusion pump. Beam current is monitored on an Eldorado Electronics microammeter with a range of .003 to 1000 microamps, model C1-1110. An Eldorado Electronics model C1-110 microcoulomb current integrator is utilized for measuring total charge accumulation for each data point.

A manifold is attached to the top of the stem of the glass tee target chamber to provide for gas inlet, pressure monitoring, and target chamber evacuation. Chamber pressure is measured by a pair of Wallace and Tiernan gauges; one gauge ranging from 0 to 50 torr and the other from 0 to 800 torr. Estimated error on these gauges is $\pm .2$ torr and ± 3 torr respectively. A quartz crystal lens with a focal length of 15 centimeters is located in the base of the tee, at an angle of 90° to the beam. The reaction spectrum immediately adjacent to the aluminum foil window is focused by this lens and falls on a 250 micron slit of a Jerral Ash monochrometer, after passing through a mechanical chopper. The Jerral Ash monochrometer has a resolving power about the central maximum of plus or minus seven angstroms with a

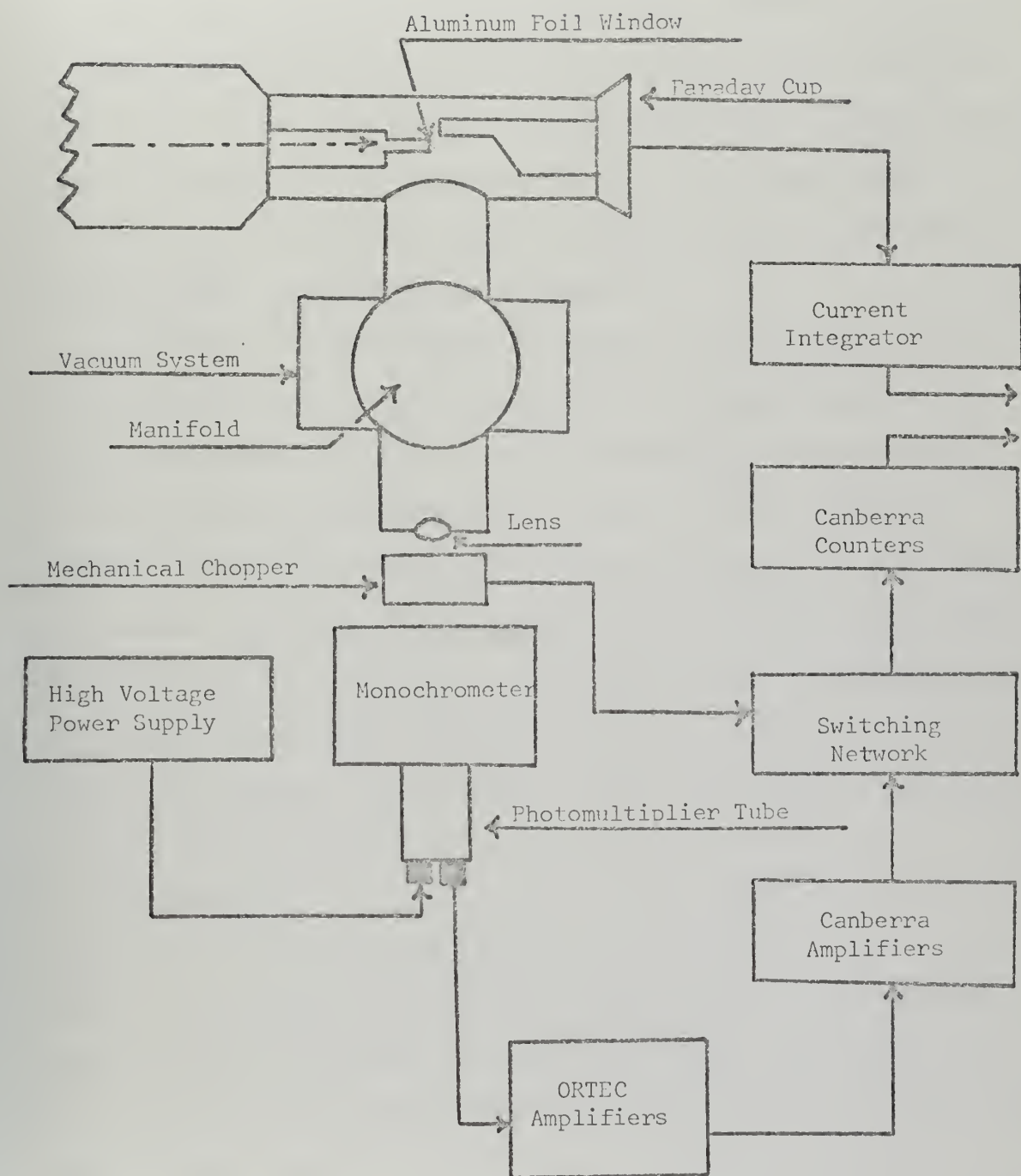


Figure 3. Schematic Diagram for Collision Chamber and Electronics.

determined error of minus 12 angstroms. The light from the spectral line under observation exits the Jerral Ash monochrometer through another 250 micron slit and is then incident on a AMPEREX XP 1110 photomultiplier tube, which has an efficiency of 20% and 38% for 5852Å and 3664Å respectively. Alignment of the detection system is checked using a neon-CO₂ laser prior to making any data run. The output signal of the photomultiplier tube is maximized through the use of a Princeton Applied Research Model HR-8 lock-in amplifier.

For each data run the electronic sequence for each pulse is from the photomultiplier tube to an Ortec Model 101 preamplifier, to a Canberra Model 810 amplifier, to a Canberra Model 830 discriminator, and finally to two Canberra Model 840 scalers. These scalers are gated by a switching circuit which is directly connected to the mechanical chopper. One scaler counts during the period light passes through the chopper and the other scaler counts when no light is allowed through the chopper, i.e., "dark" or background count. A Berkeley Model 903 double pulse generator, in conjunction with a third Canberra scaler is used as a timing circuit.

The equation

$$I = N_c / Q$$

where N_c is the total corrected photon count, and Q is the collected charge, yields the intensity of the spectral line being observed. N_c is the "light" count corrected for a system dead time of 1.7×10^{-6} sec minus the "dark" count.

For observation of neon gas without a quenching agent gas, the neon is admitted to the target chamber in increments up to a pressure of approximately 800 torr. Intensity and pressure data are recorded for each increment on each run. When utilizing a quenching gas, (nitrogen)

the neon gas is first admitted to the target chamber in increments up to a pressure of approximately 100 torr. Thereafter the partial pressure of neon is held constant and the quenching gas is admitted in increments until a total pressure of approximately 800 torr is obtained. As before, intensity and pressure data are recorded for each point of each run.

Plotting of the various curves, P/I versus I and P versus I , data reduction, and calculation of the constants A and B will be accomplished through the use of a Hewlett-Packard Model 9100 A computer.

IV. RESULTS AND CONCLUSIONS

As previously stated, due to continuous problems with the Van De Graaff accelerator, no actual reproducible data was taken. Therefore there are no conclusions as to the collisional de-excitation cross sections or collisional de-excitation reaction rates for the spectral lines of $5852.49\overset{\circ}{\text{\AA}}$ and $3664.11\overset{\circ}{\text{\AA}}$.

In attempting to obtain usable, reproducible data a run using electrons was made on N_2 . Specifically, a run was made observing the $3371\overset{\circ}{\text{\AA}}$ spectral line of N_2 to confirm the feasibility of using electrons for excitation of the target gas. Unfortunately the results using electrons for excitation were off by a factor of 10^2 to 10^3 from the results using protons for excitation, which indicates still unresolved "bugs" when using electrons for excitation.

It is hoped that successive students will carry the experiment with neon gas to fruition.

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Collisional Neons

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LINK B

LINK C

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